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Key Points:
- Natural and anthropogenic sources contributed 26% (0.64 Tg N_2O-N/year) and 74% (1.78 Tg N_2O-N/year) of the net N_2O emissions, respectively
- Pasturelands were the single largest contributor to N_2O fluxes, accounting for 86% of the net N_2O emissions (2.4 Tg N_2O-N/year)
- Among different sources, livestock excreta N deposition was the largest source of N_2O contributing to 54% of the net N_2O emissions

Supporting Information:
- Supporting Information S1

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Global Nitrous Oxide Emissions From Pasturelands and Rangelands: Magnitude, Spatiotemporal Patterns, and Attribution

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Abstract The application of manure and mineral nitrogen (N) fertilizer, and livestock excreta deposition are the main drivers of nitrous oxide (N_2O) emissions in agricultural systems. However, the magnitude and spatiotemporal variations of N_2O emissions due to different management practices (excreta deposition and manure/fertilizer application) from grassland ecosystems remain unclear. In this study, we used the Dynamic Land Ecosystem Model to simulate the spatiotemporal variation in global N_2O emissions and their attribution to different sources from both intensively managed (*pasturelands*) and extensively managed (*rangelands*) grasslands during 1961–2014. Over the study period, pasturelands and rangelands experienced a significant increase in N_2O emissions from 1.74 Tg N_2O-N in 1961 to 3.11 Tg N_2O-N in 2014 ($p < 0.05$). Globally, pasturelands and rangelands were responsible for 54% (2.2 Tg N_2O-N) of the total agricultural N_2O emissions (4.1 Tg N_2O-N) in 2006. Natural and anthropogenic sources contributed 26% (0.64 Tg N_2O-N/year) and 74% (1.78 Tg N_2O-N/year) of the net emissions, respectively. Across different biomes, pasturelands (i.e., C3 and C4) were the single largest contributor to N_2O fluxes, accounting for 86% of the net global emissions from grasslands. Among different sources, livestock excreta deposition contributed 54% of the net emissions, followed by manure N (13%) and mineral N (7%) application. Regionally, southern Asia contributed 38% of the total emissions, followed by Europe (29%) and North America (16%). Our modeling study demonstrates that livestock excreta deposition and manure/fertilizer application have dramatically altered the N cycle in pasturelands, with a substantial impact on the climate system.

1. Introduction

Nitrous oxide (N_2O) is a potent greenhouse gas (GHG), with a 100-year global warming potential 265–298 times that of carbon dioxide (CO_2, Myhre et al., 2013). The terrestrial biosphere emitted about 5.3 Tg N_2O-N in 2006 (Syakila & Kroeze, 2011) with food production contributing up to 60% of the total anthropogenic emissions (Ciais et al., 2014; Syakila & Kroeze, 2011; Tian et al., 2016). Among the anthropogenic sources (agriculture, industry, biomass burning, and indirect emissions from reactive N), agriculture plays a dominant role in driving the emission growth (Davidson, 2009; Mosier et al., 1998), contributing ~25–30% of all terrestrial biogenic emissions (Tian et al., 2016). The dominant contribution from agricultural soils is attributed to the expansion of agricultural land area and high N fertilizer use since the preindustrial era (Forster et al., 2007; Reay et al., 2012).

Nitrous oxide emissions are primarily driven by two biological processes of nitrification and denitrification (Davidson, 1991; Senbayram et al., 2009). Nitrification involves the oxidation of ammonium (NH_4^+) to nitrate with N_2O as the byproduct, while denitrification is the reduction of nitrate (NO_3^-) into dinitrogen (N_2), with N_2O as an intermediate product. These processes are regulated by various environmental factors, particularly soil water content, soil temperature, soil pH, aeration, and substrate (NH_4^+ and NO_3^-) availability (Bouwman, 1990; Dobbie et al., 1999; Granli & Bøckman, 1994; Xu et al., 2017). Although both nitrification and denitrification occur simultaneously within soils, low soil water content and coarse texture soil favor nitrification, while high soil water content and fine texture soil with high organic matter content promote denitrification (Davidson, 1991). In addition, field measurements suggest that high N_2O emissions are generally associated with soil conditions that promote denitrification (anaerobic with good NO_3^- supply, De Klein & Eckard, 2008).

Intensively managed and extensively managed grasslands have been identified as an important source of N_2O, largely due to an increase in the stocking density and changes in livestock production systems (De Klein & Eckard, 2008; Jones et al., 2005; Oenema et al., 1997). In grassland ecosystems, the major sources of N_2O emissions are livestock excreta deposition, manure N application, and mineral N application (Jones et al., 2005; Oenema et al., 1997). Livestock excreta deposition refers to all forms of livestock deposited manure N that are recycled on systems where it is produced. Manure N application refers to any forms of manure collected from the sheds and grazing lands and applied to other nearby croplands/grasslands. In particular, livestock excreta deposition, manure N application, and mineral fertilizer N application (hereafter the word *fertilizer* will be used for *mineral fertilizer*) are responsible for ~80% of the total emissions from grasslands and cropland management activities (Steinfeld, Gerber, et al., 2006). While studies are consistent toward attributing the largest sources of N_2O emissions to livestock excreta deposition, there are large differences with respect to the magnitude of N_2O emissions from different sources in intensively managed (hereafter the word *pasturelands* will be used for intensively managed grasslands) and extensively managed (hereafter the word *rangelands* will be used for extensively managed grasslands) grassland ecosystems (Davidson, 2009; De Klein & Eckard, 2008; Steinfeld, Gerber, et al., 2006). Therefore, a better understanding of the contributing factors to N_2O emissions from pasturelands and rangelands will help to reduce uncertainties in emission estimates.

The largest source of N_2O emissions in pasturelands and rangelands (see methodology section for details) comes from livestock excreta deposition and manure N application (Steinfeld, Gerber, et al., 2006). In particular, N_2O emissions associated with livestock excreta deposition in pasturelands and rangelands are six-fold higher than N_2O emissions associated with manure application to croplands (Tubiello et al., 2013). But there is considerable uncertainty regarding the contribution of different management activities (i.e., livestock excreta deposition and manure/fertilizer application) to N_2O emissions. For example, Oenema et al. (2005) found that manure N applied to soils (both croplands and grasslands) and livestock excreta deposition resulted in the emission of 0.15 and 0.62 Tg N_2O-N, respectively, in 2000. Contrastingly, Food and Agriculture Organization Corporate Statistical Database ([FAOSTAT], 2018) report that external manure N application and livestock excreta produced during grazing contributed to an emission of 0.35 and 1.46 Tg N_2O-N, respectively, in the same year. These large differences in the estimates of N_2O emissions from manure N are associated with (1) differences in animal categorization and N excretion rates per animal species, (2) partitioning of total manure N production to livestock manure deposition on pasturelands and rangelands, (3) estimates of N lost through leaching, (4) allocation of total manure application to grasslands and croplands, and (5) the spatial and temporal resolution of data used to estimate emissions (Bouwman et al., 1995).

Likewise, N_2O fluxes in grassland systems can differ as a function of plant community composition (C3, C4, and mixed species; Mosier et al., 1991; Tiemann & Billings, 2008). Plant functional type (PFT) and differences in community composition alter soil N_2O fluxes, particularly through their effect on soil N availability. Perennial plants have a longer growing season and are expected to leave less N in soils for N_2O conversion (Oates et al., 2016). Likewise, mixed species plots have been found to emit significantly lower N_2O from soils compared to either C3 or C4 plots (Epstein et al., 1998), indicating that high species richness can lower N_2O emissions due to greater nutrient use by plants (Hooper & Vitousek, 1997; Tilman et al., 1997), resulting in lower soil N available for nitrification/denitrification. Plant community composition also affects N_2O fluxes due to changes in litter quality and C:N ratio, with higher N_2O fluxes under reduced litter C:N ratios (Wedin & Tilman, 1996). For example, Nichols et al. (2016) found no difference in the emission factor between C3 and C4 plants following urine or feces N deposition. In contrast, total N_2O fluxes were greater from C4 compared to C3 crop when treated with N fertilizer (Bronson & Mosier, 1993; Mosier et al., 1986). Differences in N_2O emissions between C3 and C4 plants are likely due to differences in N turnover associated with differences in litter quality or C:N ratios and the ability of C3 plants to recover or retain more N following N application (Mosier et al., 1986). But Epstein et al. (1998) suggest that plant community composition can play an essential role only when soil moisture and temperature are not limiting factors, indicating that the response of N_2O fluxes to C3 versus C4 grass can be regulated by soil moisture and temperature changes.

Process-based models with explicit representation of nitrification and denitrification mechanisms are valuable tools for constraining estimates of N_2O emissions and attributing those emissions to different sources including climate and management practices (Bouwman et al., 2002; Lu & Tian, 2013; Stehfest & Bouwman, 2006; Tian, Chen, et al., 2015). Chamber and tower-based approaches are useful in estimating N_2O emissions at the site-level, but high temporal and spatial variations in N_2O fluxes to multiple environmental factors and land management practices, make it difficult to extrapolate site-based estimates to regional and global scales (Smith et al., 2008). Process-based models that simulate N_2O emissions as a function of seasonal fluctuation in soil temperature, soil moisture, and resource (ammonium and nitrate) availability are becoming increasingly valuable for estimating N_2O emissions at large scales and predicting the influence of management activities and climate change on N_2O emissions (Tian et al., 2018).

There are considerable differences among process-based models for the simulation of livestock excreta deposition and manure/fertilizer N application. Some models have an explicit representation of manure N production by animals (Dangal, Tian, Lu, et al., 2017), while other models assume manure to be an external prescribed model input. For example, the Integrated Farm System Model and the Global Livestock Environmental Assessment Model simulate all farm components (herd structure; production and turnover of manure and their application to feed crops; direct and indirect energy use; the composition of ration for each species and production system; energy requirement for each animal cohort; production of meat, milk, and eggs; and environmental impacts of production) to provide estimates of GHG fluxes at the farm level (Gerber et al., 2013; Rotz et al., 2012). In contrast, other models such as DayCent use fertilizer and manure N data as model inputs (Bouwman et al., 2013; Li et al., 1992; Parton et al., 1996; Schmid et al., 2001). Processes following N input to the system include internal recycling of N in the grassland where it is produced, displacement and recycling in another cropland or grassland, usually in the same region, volatilization, leaching, and dung burning losses (Oenema et al., 2005). Models also show large differences in N_2O fluxes in response to livestock excreta deposition or manure N application, with relative deviations of +38% (flux difference = 1 kg N_2O-N/ha) to +258% (flux difference = 2.58 Kg N_2O-N/ha) compared to observations during 2002–2004 (Abdalla et al., 2010). The overestimation of simulated N_2O fluxes compared to observation was attributed to large soil water filled pore space (WFPS) that favors high denitrification rates. While accurately simulating soil moisture dynamics and WFPS is critical to estimate N_2O emissions, differences in model structure, sources of N input data, and microbial responses to manure application/deposition (Wennman & Kätterer, 2006) can further add large uncertainty. Likewise, previous modeling studies were limited to sites and regional scales (Gerber et al., 2013; Soussana et al., 2007; Wolf et al., 2010) and also failed to consider all forms of N while estimating N_2O emissions from pasturelands and rangelands. In addition, although grazing livestock are responsible for manure N deposition of 65.9 Tg N annually between 1961 and 2016 (FAOSTAT, 2018), spatially explicit estimates of N_2O fluxes are lacking at the global scale. In this study, we aim to reduce uncertainties in the estimates of N_2O emissions from pasturelands and rangelands by (1) considering all forms of N inputs using the integrated framework, (2) attributing N_2O emissions to different sources including livestock excreta deposition and manure/fertilizer applications, and (3) quantifying regional and biome specific variations in N_2O fluxes to locate hot spots of N_2O emissions. To accomplish this task, we use a highly integrated land model (The Dynamic Land Ecosystem Model, DLEM) to examine the response of N_2O emissions to climate and management intensity. The model has been applied widely to estimate preindustrial N_2O emissions (Xu et al., 2017) and contemporary GHG (CH_4, N_2O, and CO_2) fluxes (Tian, Xu, et al., 2010; Tian, Chen, et al., 2015) from terrestrial ecosystems at regional to global scale.

We hypothesize that (i) pasturelands would account for a significant proportion of global agricultural N_2O emissions, largely due to an increase in excreta N production as a result of higher stocking rates and an increase in live weight of animals over time, driven by human population growth, urbanization, and rising incomes (Thornton, 2010); (ii) high precipitation would likely have a positive effect on soil moisture dynamics and WFPS creating anaerobic condition, that together with excreta and manure/fertilizer N input, would lead to an increase in N_2O emissions. However, the effects of high precipitation can be modified by soil conditions, with relatively high N_2O emissions from fine texture soil due to easily reachable anaerobic

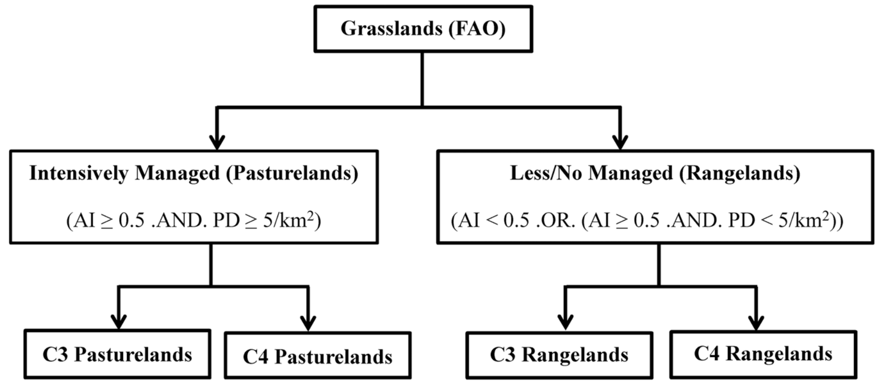

Figure 1. Overall workflow used to categorize grasslands in this study. Gridded grasslands spatial distribution maps are based on Klein Goldewijk et al. (2017), which were allocated to match FAO country level grassland categories *permanent meadows and pastures*. Pasturelands and Rangelands were further categorized into C3 and C4 types by overlaying the global distribution of C3 and C4 grasslands based on Still et al. (2003). AI = aridity index; PD = population density; FAO = Food and Agriculture Organization.

conditions that are further extended for longer periods compared to coarse texture soils (Bouwman et al., 2002); (iii) warming would increase N_2O emissions due to an increase in the rates of enzymatic processes in soils, provided that other factors (e.g., substrate or moisture supply) have no limiting effect (Schindlbacher et al., 2004); (iv) excreta N deposition would account for the majority of N_2O fluxes because excreta N deposition is several orders of magnitude higher than manure/fertilizer N application in pasturelands and rangelands (FAOSTAT, 2018); and (v) different plant functional types (C3 and C4) would likely result in variation in N_2O fluxes due to differences in litter quality and C:N ratios. In addition, since the photosynthetic rate per unit N is lower in C3 compared to C4 plants (Kellogg, 2013; Sage & Pearcy, 1987), C3 plants are able to recover, retain, and use more N following N application compared to C4 plants (Mosier et al., 1986). But such effect can be modified by temperature, soil moisture changes, and the nutrient limitation at the site (Epstein et al., 1998; Felzer et al., 2011).

2. Materials and Methods

2.1. Definition of Grasslands

A variety of definition exists for grasslands (see Text S1 and Figure S1 in the supporting information). Globally, grassland ecosystems are defined as areas dominated by herbaceous and shrub vegetation, which includes savannas (Africa, South America, and India), steppe (Eurasia), prairies (North America), shrub-dominated areas (Africa), meadows (United Kingdom and Ireland), and tundra (Breymeyer, 1990; Graetz, 1994; White et al., 2000). In this study, we defined grasslands as areas covered by *permanent meadows and pasturelands* or *grazing land* (FAOSTAT, 2018) and used by Klein Goldewijk et al. (2017) for reconstructing historical agricultural lands in the HYDE3.2 land use dataset (Figure 1). Under the Food and Agriculture Organization (FAO) definition, permanent meadows and pastures include land under permanent use (for five consecutive years or more) to grow herbaceous forage either through cultivation or naturally. Klein Goldewijk et al. (2017) used satellite maps of herbaceous land cover from ESA-CCI for the year 2010, adjusted such that the area of managed grasslands matches the area of country level FAO categories permanent meadows and pastures or grazing land. Klein Goldewijk et al. (2017) further categorized permanent meadows and pastures into intensively managed (pasturelands) and extensively managed (rangelands) as a function of aridity index and population density. Aridity index measures the degree of water stress at a given location and is estimated as a ratio of mean annual precipitation and mean annual evapotranspiration. A grid is categorized as rangelands when the aridity index is less than 0.5 or when the aridity index is higher than 0.5 but the population density is less than 5/km^2.

Following the development of global pasturelands and rangelands distribution data, we (1) simulated the evolution of N_2O emissions from pasturelands and rangelands during 1961–2014; (2) quantified biome-specific and regional variations in N_2O fluxes from pasturelands and rangelands; (3) attributed trends in simulated N_2O emissions to changes in climate and grassland management intensity, the later including livestock excreta deposition and manure/fertilizer N application; and (4) evaluated the impacts of precipitation and temperature changes on N_2O emissions from pasturelands and rangelands.

2.2. The DLEM

The DLEM is a process-based ecosystem model that provides estimates of the carbon, N, and water stocks and fluxes in land ecosystems using spatially explicit information on multiple environmental factors including vegetation, soil (texture, bulk density, and pH), climate (mean, maximum and minimum air temperatures, and precipitation), and land management practices (fertilization and manure application) at different scales (Pan, Tian, et al., 2015; Tian, Chen, et al., 2015; Tian et al., 2012). The biophysical and geochemical processes in the DLEM are simulated through the interaction of five core components, which includes biophysics, plant physiology, soil biogeochemistry, dynamic vegetation and disturbance, and land use and land management practices. The detailed description of the model structure, algorithm, and parameterization has been documented elsewhere (Pan, Dangal, et al., 2015; Pan et al., 2014; Ren et al., 2012; Tian, Xu, et al., 2010; Tian, Chen, et al., 2010; Tian, Yang, et al., 2015). In this study, we improved the DLEM (version 2.0) to account for N fertilizer application and manure deposition, and to make estimates of N_2O emissions from pasturelands and rangelands during 1961–2014.

2.3. Model Improvements

In the previous version of the DLEM 2.0, management practices including N fertilizer application and irrigation were only implemented for croplands, while grasslands were broadly categorized as C3 and C4 grasses and treated as natural biome (Lu & Tian, 2013; Pan et al., 2014; Ren et al., 2007; Tian, Xu, et al., 2010). In this version, we improved the model by representing processes for four different grassland types: (1) C3 pasturelands, (2) C4 pasturelands, (3) C3 rangelands, and (4) C4 rangelands. We assumed that pasturelands experience livestock excreta deposition and applications of fertilizer and manure N, while rangelands experience livestock excreta deposition only. Management intensity (excreta deposition, manure, and fertilizer application) was prescribed in the model as an external input. Details on carbon and N cycling in and out of the pasturelands and rangelands are available in Text S2 (Vaddella et al., 2010). N inputs in the soil occur through processes such as atmospheric N deposition, livestock manure deposition, and manure/fertilizer application, while N outputs occur through processes such as plant uptakes, runoff and leaching, and N-containing gas emissions. The availability of substrate (inorganic N) plays an important role in determining the nitrification and denitrification processes (Chatskikh et al., 2005), which can be further modified by soil temperature and moisture changes (Kirschbaum, 1995; Li et al., 2000; Lin et al., 2000; Petersen et al., 2005, Texts S2 and S3). Following nitrification and denitrification, N_2O is separated from NO and N_2 using an empirical equation reported by Davidson et al. (2000, Text S3). These processes are assumed to occur only in the top 50-cm soil surface.

We also accounted for differences in plant community composition (C3, C4, or mixed) while simulating N_2O emissions from pasturelands and rangelands. The DLEM simulates carbon assimilation rates of C3 and C4 grasses as a minimum function of three limiting factors: (a) photosynthetic enzyme (rubisco), (b) photosynthetically active radiation (light), and (c) photosynthetic production utilization (export, Collatz et al., 1991; Farquhar et al., 1980). In the case of C4 species, the export limitation refers to the phosphoenolpyruvate carboxylase limited rate of assimilation. Total assimilated carbon is then allocated to leaves, stems, and roots as a function of PFT-dependent allocation rates (Table S1), which further determines the amount of carbon and N entering the soil (litterfall), after accounting for losses through autotrophic respiration (Friedlingstein et al., 1999). The amount and type of inorganic N in soil regulate nitrification and denitrification processes (Texts S4 and S5), which eventually determine the production and fluxes of N_2O from the soil (Bijay-Singh et al., 1989; Chatskikh et al., 2005; Chowdhury et al., 2017; Fortuna et al., 2003; Zhang, Peng, et al., 2017).

2.4. Input Data Sets

The input data and simulation protocol are generally consistent with the Nitrogen Model Intercomparison Project (Tian et al., 2018). Here we provide a brief description of these data sets and modified simulation protocol associated with this study. Gridded, georeferenced data sets for the DLEM were compiled from various sources at a spatial resolution of 0.5° × 0.5°. These data sets include daily climate data, atmospheric

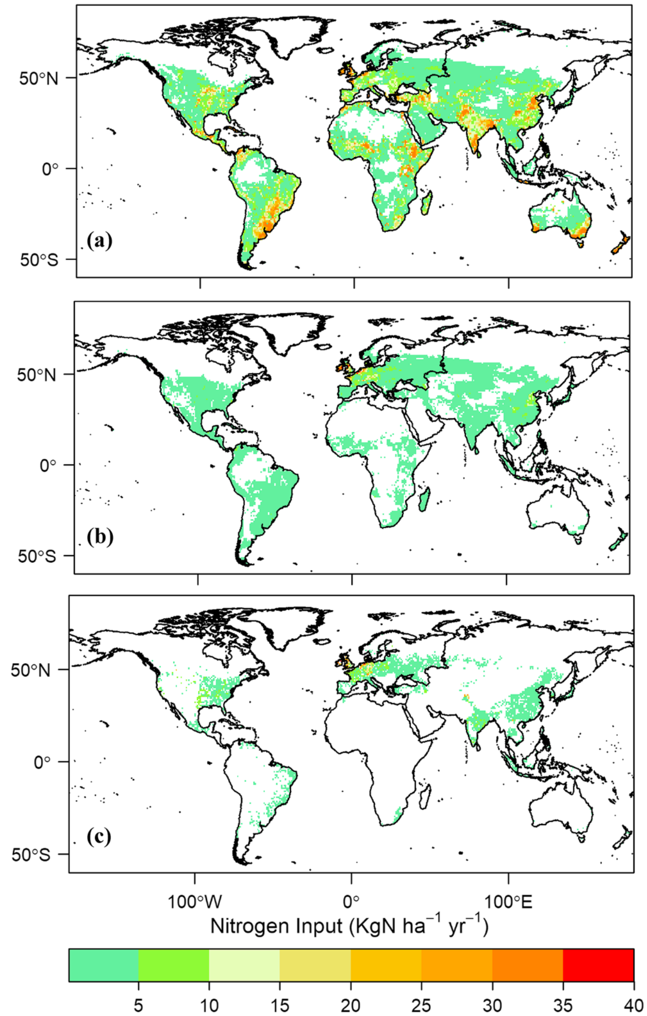

Figure 2. N input in pasturelands and rangelands associated with excreta N deposition (a), manure N application (b), and N fertilizer application (c) during 1961–2014.

chemistry (CO_2 concentration, AOT40 O_3 index, and N deposition), soil properties, land management practices (excreta N deposition, manure N application, and fertilizer N application), and other ancillary data such as river network, cropping system, and topography maps. Daily climate data during 1901–2014 were based on Climatic Research Unit-National Centers for Environmental Prediction (CRU-NCEP) climate forcing (ftp://nacp.ornl.gov/synthesis/2009/frescati/model_driver/cru_ncep/analysis/readme.htm), while atmospheric CO_2 concentration was obtained from Carbon Dioxide Information Analysis Center (http://cdiac.ornl.gov/). Annual N deposition was retrieved from the outputs of multiple atmospheric chemistry transport models (http://daac.ornl.gov/CLIMATE/guides/global_N_deposition_maps.html; Dentener et al., 2006), and tropospheric ozone concentration was based on Felzer et al. (2004), while annual changes in pasturelands and rangelands were based on Klein Goldewijk et al. (2017). Elevation, slope, and aspect were derived from Global 30-Arc Second Elevation Product (https://lta.cr.usgs.gov/GTOPO30), and soil texture was derived from FAO Soil Database System (Reynolds et al., 2000). N input to global pasturelands and rangelands are based on Xu et al. (2019), where excreta N deposition was developed by integrating country level Food and Agriculture Organization Corporate Statistical Database (FAOSTAT) database on *manure left on pastures* with gridded total manure production data sets based on Zhang, Tian, et al. (2017; Figures 2a and S2). Similarly, manure N application was developed by combining country level FAOSTAT database on *manure applied to soils* with the ratio of total pasturelands area to total agricultural area (croplands and pasturelands, Figures 2b and S2). Gridded N fertilizer application in pasturelands was obtained by combining the ratio of N use on pasturelands to total fertilizer N input in 151 countries based on Lassaletta et al. (2014) with HYDE pasturelands area during 1961–2014 (Figures 2c and S2). Global livestock production had resulted in a net excreta deposition of 67 Tg N/year, with C3 and C4 pasturelands accounting for nearly half of the total excreta N production during 1961–2014. The net N input following manure application is about 7.3 Tg N/year, where 89% of the manure is applied to C3 pasturelands. Similarly, N input following fertilizer application is about 4.2 Tg N/year, where 93% of the fertilizer is applied to C3 pasturelands.

Pasturelands and rangelands input data are based on HYDE 3.2 (Klein Goldewijk et al., 2017), which assumes that pasturelands are found closer to populated areas and are representative of wetter grazing lands, while rangelands are found in drier areas. The global distribution of pasturelands and rangelands for 2014 is provided in Figure S3. We further categorized pasturelands and rangelands into C3 and C4 categories by overlaying the global distribution of C3 and C4 grasslands based on Still et al. (2003, Figure S4). Still et al. (2003) used crossover temperature approach to classify grid cells as favorable to C3 and C4 species using the climate data. After classifying the grid cells as C3 and C4 species, the fraction of vegetation that could be C4 is estimated using information on the distribution of woody and herbaceous vegetation. Since nearly all C4 species are restricted to the herbaceous growth form, Still et al. (2003) assumed that the herbaceous area is the maximum area occupied by C4 species in a particular grid cell. However, it is important to recognize that grassland planted with C4 species is likely not included in Still et al. (2003) data sets because C4 species distribution was driven by climate information only.

2.5. Experimental Design

For each grid cell, we first run the DLEM to determine the equilibrium state of carbon, N, and water using the early twentieth century (30 years; 1901–1930) daily climate, while other input data (atmospheric CO_2, N deposition, and land cover) were kept at 1900 level. To determine the equilibrium state for each grid cell, the

Table 1
Simulation Design Used in This Study and the Respective Mean N_2O Fluxes Associated With Different Simulations

	Simulation	Climate	CO_2	O_3	NDEP	Fertilizer application	Manure application	Excreta deposition	N_2O fluxes (Tg N_2O-N/year)
S0	Reference	30-year average[a]	1901	1901	1901	1901	1901	1901	0.46
S1	Multifactor	1901–2014	1901–2014	1901–2014	1901–2014	1961–2014	1901–2014	1901–2014	2.43
S2	No Man./Fert.	1901–2014	1901–2014	1901–2014	1901–2014	NA	NA	NA	0.64
S3	Climate only	1901–2014	1901	1901	1901	1901	1901	1901	0.01
S4	Fertilizer App.	30-year average	1901	1901	1901	1901–2014	1901	1901	0.17
S5	Manure App.	30-year average	1901	1901	1901	1901	1901–2014	1901	0.33
S6	Excreta Dep.	30-year average	1901	1901	1901	1901	1901	1901–2014	1.31

[a]Indicate the 30-year daily mean climate value from 1901 to 1930. Reference simulation (S0) is used to estimate preindustrial N_2O emissions, while multifactor include simulation with historical changes in climate, carbon dioxide (CO_2), tropospheric zone (O_3), atmospheric N deposition (NDEP), and land management practices (fertilizer N, manure N, and exrecta deposition). No Man./Fert. (S2) is a simulation in the absence of nitrogen input in the form of excreta deposition, manure application, and fertilizer application. Climate (S3) is a simulation with transient climate over the study period, but all other factors are kept constant at 1901 level. Fertilizer app. (S4), manure app. (S5), and excreta dep. (S6) are simulation with 30-year daily mean climate and transient fertilizer application, manure application, and excreta deposition, respectively; but all other factors are kept constant at 1901 level. The tenth column represent the mean N_2O fluxes based on different model simulations during 1961–2014.

DLEM was allowed to run for the maximum of 10,000 years or until the change in carbon fluxes is less than 0.5 g C/m^2/year, the change in soil water pool is less than 0.5 mm/year, and the change in total N content is less than 0.5 g N/m^2/year during two consecutive 20 years. Following the equilibrium run, the model spin up was carried to allow for smooth transition from equilibrium to transient state by randomly selecting 20 years of climate data, repeated 5 times, during 1901–1930.

Following a model spin up, we performed seven (S0–S6) simulations to determine the magnitude and relative contribution of climate and land management practices (excreta deposition, manure application, and fertilizer application; Table 1). The S0 (reference run) is carried out with daily mean climate data (1901–1930) and other environmental forcings at 1901 level. The purpose of S1 (Multifactor) simulation is to quantify the overall contribution of multiple environmental changes and land management practices on N_2O emissions. S0 is subtracted from S1 to estimate the overall contribution of multiple environmental changes on N_2O emissions during 1961–2014. The simulation S2 (No Man./Fert.) is designed to separate anthropogenic contribution to N_2O emissions and is carried out in the absence of excreta deposition, manure application, and fertilizer application with transient climate, CO_2, O_3, and nitrogen deposition. The anthropogenic contribution in this study is defined as N_2O fluxes arising from manure N and fertilizer N application in pasturelands, and excreta N deposition in both pasturelands and rangelands. The anthropogenic sources are estimated as the difference between S1 and S2 simulation. The rest of the simulations (S3–S6) are designed to quantify the contribution of individual factors (climate, fertilizer N application, manure N application, and excreta N deposition) on N_2O emissions. To assess the influence of temperature and precipitation on N_2O fluxes, we developed linear regression model using annual N_2O fluxes from the S1–S2 simulation as a response variable and annual mean temperature and precipitation totals as an explanatory variable. We used outputs from S1–S2 scenario to assess the relationship between temperature/precipitation and N_2O fluxes because S1–S2 simulation includes the influence of climate and its interaction with other environmental factors and land management practices.

2.6. Statistical Analysis and Uncertainty Estimates of N_2O Fluxes

We used mean and 1-sigma standard deviation to provide estimates of N_2O emissions and their uncertainty during the study period. To test the statistically significant trend in N_2O fluxes during 1961–2014, we used nonparametric Mann-Kendall method (Kendall, 1975; Mann, 1945). In addition, the relationship between climate, land management activities, and N_2O fluxes was assessed by performing linear regression analysis using R (https://www.r-project.org).

To estimate the uncertainty in N_2O fluxes due to uncertainty in N input data, we first determine the temporal change in N input from excreta deposition, manure application and fertilizer application, and their associated N_2O fluxes. We then assess the relationship between N_2O fluxes and N input using bootstrapping method with 10,000 replicates to determine the 95% confidence interval of the slope of that relationship. We

Table 2
Comparison of DLEM-Simulated N_2O Emission Against Observations in Rangelands and Pasturelands

Site (Lat/Lon)	Year	Vegetation	Manure/fertilizer type	Nitrogen levels (kg N/ha)	Measured (kg N_2O-N/ha)	Modeled (kg N_2O-N/ha)	References
10°30′S, 62°30′W	2001	C4 Pastureland	NA	0	0.07	0.08	Carmo et al. (2005)
51°46′N, 9°42′E	2009	C3 Rangeland	NA	0	0.51	0.4	Hoeft et al. (2012)
40°50′N, 104°42′W	1997–2000	C4 Rangeland	NA	0	0.10	0.14	Mosier et al. (2002)
42°59′N, 2°37′W	1998	C3 Pastureland	NA	0	0.32	0.49	Merino et al. (2002)
			Manure	80	1.24	0.77	
			Manure	85	1.23	0.79	
55°53′N, 3°26′W	1992	C3 Pastureland	NA	0	0.04–0.26	0.02	Clayton et al. (1997)
			Fertilizer	360	0.69–1.28	1.01	
			Manure	360	0.48–6.39	1.74	
45°33′N, 116°42.3′E	2007–2008	Typical steppe	NA	0	0.22 ± 0.07	0.20	Wolf et al. (2010)[a]

Note. DLEM = Dynamic Land Ecosystem Model.
[a]Used ungrazed site to compare the fluxes with DLEM simulated N_2O emission for year 2007.

further determine the uncertainty in N input data (excreta deposition, manure application, and fertilizer application) using the range of annual N inputs during 1961–2014. We then created 10,000 bootstrap samples using the range of annual N inputs and estimated the 95% confidence interval for three different nitrogen forms. The 95% confidence interval of the slope is then multiplied by the uncertainty range of the N input data to estimate the uncertainty in N_2O emissions associated with excreta deposition, manure application, and fertilizer application. The bootstrapping method employed in this study relies on random sampling with replacement technique to estimate the sampling distribution of a given statistic. The bootstrapping was done using two functions (*boot and boot.ci*) in R (Canty, 2002).

2.7. Model Evaluation at the Site Level

We compared the simulated N_2O emissions for pasturelands and rangelands at different levels of N application across several sites located in South America, Europe and Inner Mongolia, China. Our results show that the DLEM-simulated N_2O fluxes are in agreement with observations, particularly for rangelands (Table 2). In case of pasturelands, DLEM showed a reasonable agreement with N_2O emissions at different levels of fertilizer and manure N applications in Edinburgh (Clayton et al., 1997) and Basque County, Spain (Merino et al., 2002), but there was a tendency of underprediction in other sites (Table 2). In Inner Mongolia, we compared simulated N_2O emissions in the absence of N input against a typical steppe site, which was ungrazed since 1999. The DLEM simulation underpredicted N_2O emissions compared to Wolf et al. (2010) by 9% (0.22 vs 0.2 Kg N_2O-N/ha). The slight underprediction compared to Wolf et al. (2010) is likely because we did not account for grazing effect on N_2O emissions prior to 1999.

We also compared N_2O emissions from pasturelands and rangelands across 12 sites in Europe, spanning a wide range of climatic, environmental, and soil conditions (Flechard et al., 2007; Table 3). The sites were classified as intensively or extensively managed primarily based on whether the site received N input in the form of fertilizer or not. N_2O emissions were reported for 3 years in Flechard et al. (2007); however, we only selected years with the highest number of measurement records while evaluating DLEM performance against these sites (Table 3). In general, DLEM overpredicted N_2O emissions at 8 out of 12 sites, largely due to differences in the amount of N input in the form of fertilizer and manure. For example, at an intensive site in Ireland (Ei-CA), DLEM showed a large deviation when compared to observation (2.47 vs 4.7 Kg N_2O-N/ha/year) because N input in the DLEM was higher than Flechard et al. (2007) by 23%. In addition, most of these sites were grazed and cut several times a year, which likely explain the discrepancies between simulated fluxes and observations.

3. Results

3.1. Temporal Patterns of N_2O Emissions From Pasturelands Versus Rangelands

The DLEM-simulated results showed that the influence of multiple environmental factors including land management practices, such as livestock excreta deposition, and manure/fertilizer N application on

Table 3
Comparison of DLEM-Simulated N_2O Emissions Against Measurements Based on Flechard et al. (2007) in Europe for Intensively and Extensively Managed Grasslands

Site	Latitude	Longitude	Country	Management intensity	Site N input (kg N/ha/year)	Observed N_2O (kg N_2O-N/ha/year)	Model N input (kg N/ha/year)	Model N_2O (kg N_2O-N/ha/year)
Hu-BGc	46°41′30″N	19°36′06″E	Hungary	Extensive	NA	0.8	NA	1.6
Hu-BGg	46°41′30″N	19°36′06″E	Hungary	Extensive	NA	0.8	NA	1.6
UK-BS	55°52′N	3°2″W	Scotland	Intensive	200	3.69	85	2.5
Ei-CA	52°51′59″N	6°54′30″W	Ireland	Intensive	200	2.47	245	4.7
UK-CP	55°52′N	3°12″W	Scotland	Extensive	NA	0.33	NA	0.6
Fr-Lae	45°38′35″N	2°44′9″E	France	Extensive	NA	0.17	NA	0.12
Fr-lai	45°38′35″N	2°44′9″E	France	Intensive	175	0.8	120	0.97
Ni-LE	52°30′N	5°30′E	Netherland	Intensive	300	3.5	365	4.9
Dk-LV1	55°41′40″N	12°07′07″E	Denmark	Intensive	200	0.28	225	0.4
It-MA	46°07′00″N	11°42′10″E	Italy	Extensive	90	0.01	NA	0.02
CH-Oee	47°17′N	7°44′E	Switzerland	Extensive	NA	0.2	NA	0.3
CH-Oei	47°17′N	7°44′E	Switzerland	Intensive	200	1.1	149	1

Note. We only used years with highest number of measurement records during the comparison. DLEM = Dynamic Land Ecosystem Model.

pasturelands and rangelands resulted in a significant increase in N_2O emissions from 1.74 in 1961 to 3.11 Tg N_2O-N in 2014, that is a linear rate of 0.027 Tg N_2O-N/year^2 ($p < 0.05$). Both pasturelands and rangelands experienced a significant increase in N_2O emissions at the rate of 0.025 and 0.002 Tg N_2O-N/year^2 during 1961–2014 ($p < 0.05$; Figure 3), respectively. In the previous 54 years (1961–2014), pasturelands contributed to 86% (2.1 Tg N_2O-N/year), while rangelands contributed to 14% (0.33 Tg N_2O-N/year) of the total emissions.

3.2. Spatial, Regional and Biome Level Variations in N_2O Emissions

Our results showed a large spatial variation in pasturelands N_2O emissions as a result of multiple environmental changes and grassland management practices such as livestock excreta deposition, and manure/fertilizer N application (Figure 4). Spatial pattern of N_2O emissions indicate that Southern Asia, Europe, North America, Africa, South America and Northern Asia experienced a significant increase in N_2O emissions ($p < 0.05$) during 1961–2014. In particular, Europe experienced a significant increase in N_2O emissions until 1990 at the rate of 0.012 Tg N_2O-N/year^2 ($p < 0.05$), but a significant decline afterward at the rate of 0.005 Tg N_2O-N/year^2 ($p < 0.05$). Southern Asia dominated N_2O emissions contributing to 38% of the total emissions, followed by Europe (29%), North America (16%) and Africa (12%) during 1961–2014 (Figure 5). The contribution from Southern Asia has increased significantly since 1961 at the rate of 0.015 Tg N_2O-N/year^2 ($p < 0.05$).

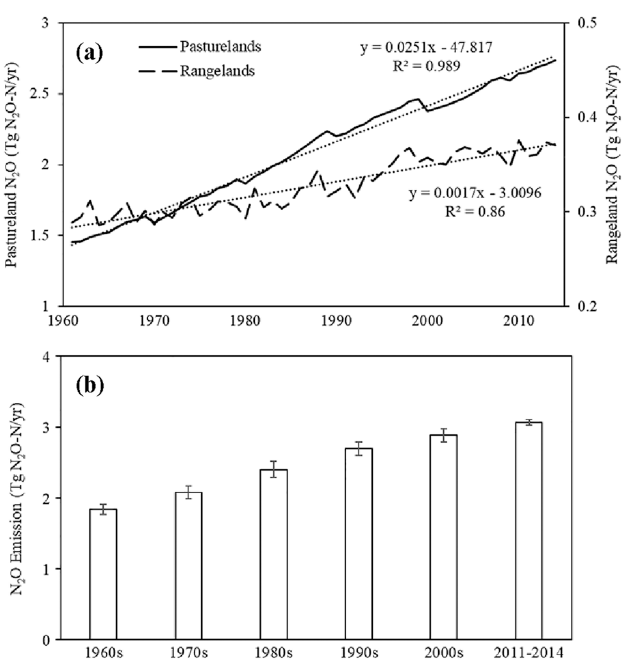

Figure 3. Temporal change in N_2O emissions from pasturelands and rangelands (a), and decadal changes in N_2O emissions from global pasturelands and rangelands (b).

Across different biomes (pasturelands and rangelands), model results showed that C3 pasturelands were the dominant source of N_2O emissions, contributing to a net flux of 1.99 Tg N_2O-N/year during 1961–2014 (Figure S5). Similarly, C3 rangelands contributed to a net N_2O flux of 0.26 Tg N_2O-N/year, followed by C4 pasturelands (0.10 Tg N_2O-N/year) and C4 rangelands (0.06 Tg N_2O-N/year). C3 pasturelands were the major source of N_2O fluxes accounting for 82% of the global emissions from pasturelands and rangelands. For specific biomes, model results indicated that C3 pasturelands experience the largest magnitude of increase in emission intensity (emissions per unit area) from 2.84 in the 1960s to 3.95 kg N_2O-N/ha/year during 2010–2014 (Figure S6). Interestingly, the comparison of relative percentage change in emission intensity between the 1960s and 2011–2014 showed that C4 pasturelands experienced the largest rate of increase in emission intensity of 62%, followed by C3

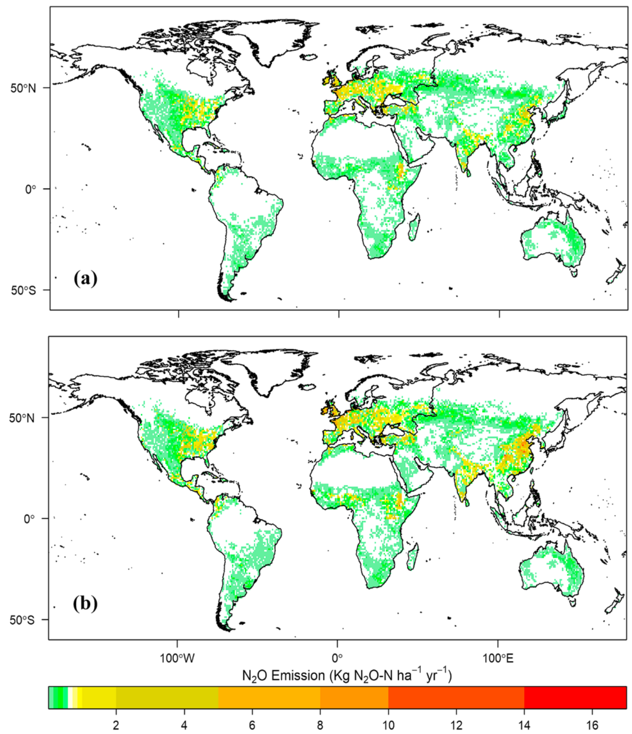

Figure 4. Spatial pattern of N_2O emissions in the 1960s (a) and the 2010s (b) as a result of multiple environmental changes. The results are based on multifactor simulation (S1) during 1961–2014.

pasturelands (39%). Regionally, Southern Asia dominated N_2O fluxes contributing to 40% of the total emissions, followed by Europe (34%), North America (15%), Africa (9%), South America (1%), and Northern Asia (1%) from C3 pasturelands (Figure 6a). Likewise, Southern Asia contributed to 36% of the net N_2O emissions from C3 rangelands, followed by North America (27%), Africa (20%), South America (6%), Oceania (5%), Europe (4%), and Northern Asia (2%, Figure 6b). In C4 pasturelands, Africa was the dominant source of N_2O fluxes, accounting for 48% of the total emissions, followed by Southern Asia (25%), South America (15%), North America (11%), and Europe (1%, Figure 6c). In C4 rangelands, Africa contributed to 41% of the total N_2O emissions, followed by Oceania (33%), South America (18%), North America (6%), and Southern Asia (2%) (Figure 6d).

3.3. Contributing Factors to Total N_2O Emissions and Emissions Per Unit N Input

Of the total N_2O fluxes, natural and anthropogenic sources contributed to 0.64 (26%) and 1.78 Tg N_2O-N/year (74%), respectively, during 1961–2014. Among the different sources of N_2O emissions, livestock excreta N deposition on pasturelands and rangelands contributed to 54% (1.31 Tg N_2O-N/year), while manure N application and fertilizer N application contributed to 13% (0.33 Tg N_2O-N/year) and 7% (0.17 Tg N_2O-N/year) of the mean N_2O emissions during 1961–2014, respectively. In pasturelands, livestock excreta deposition was the largest emission source (1.28 Tg N_2O-N/year), followed by manure N application (0.33 Tg N_2O-N/year) and fertilizer N application (0.18 Tg N_2O-N/year, Figure 7) during 1961–2014. Estimates of emission factor as the ratio of the difference between N_2O emissions from S1 and S2 simulations and total N input showed a net emissions of 0.02 Tg N_2O-N/Tg N.

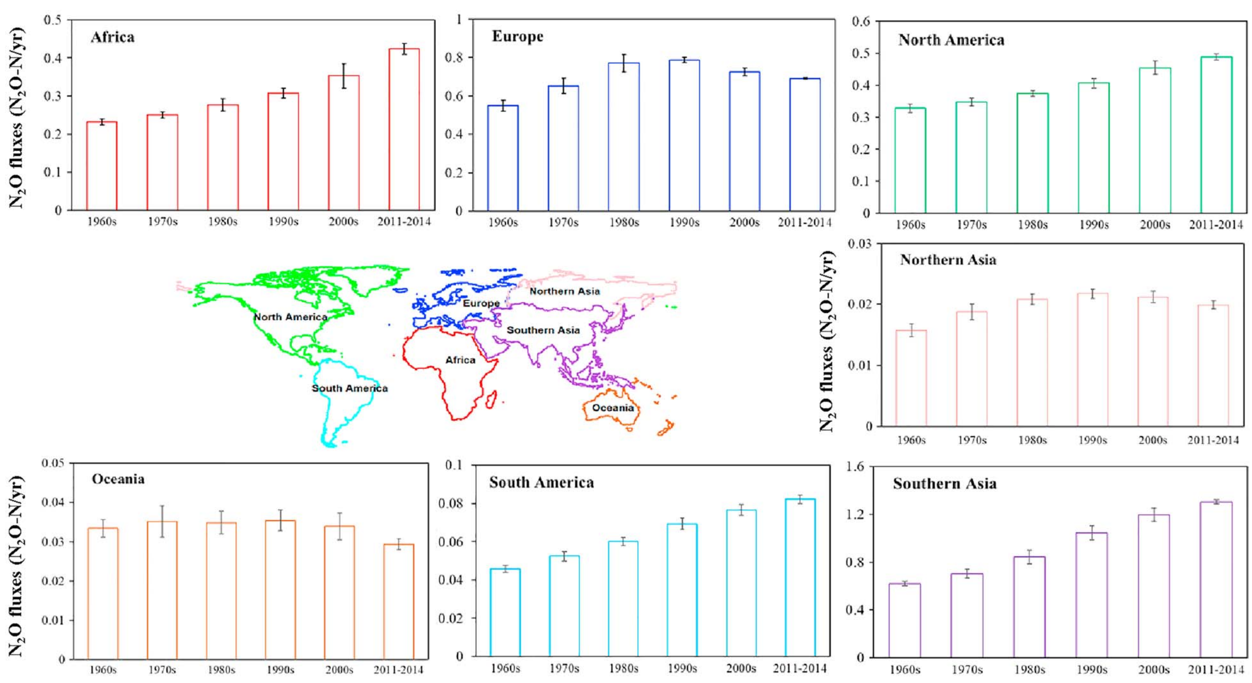

Figure 5. Regional changes in N_2O emissions during the 1960s to the 2010s based on multifactor simulation (S1) during 1961–2014.

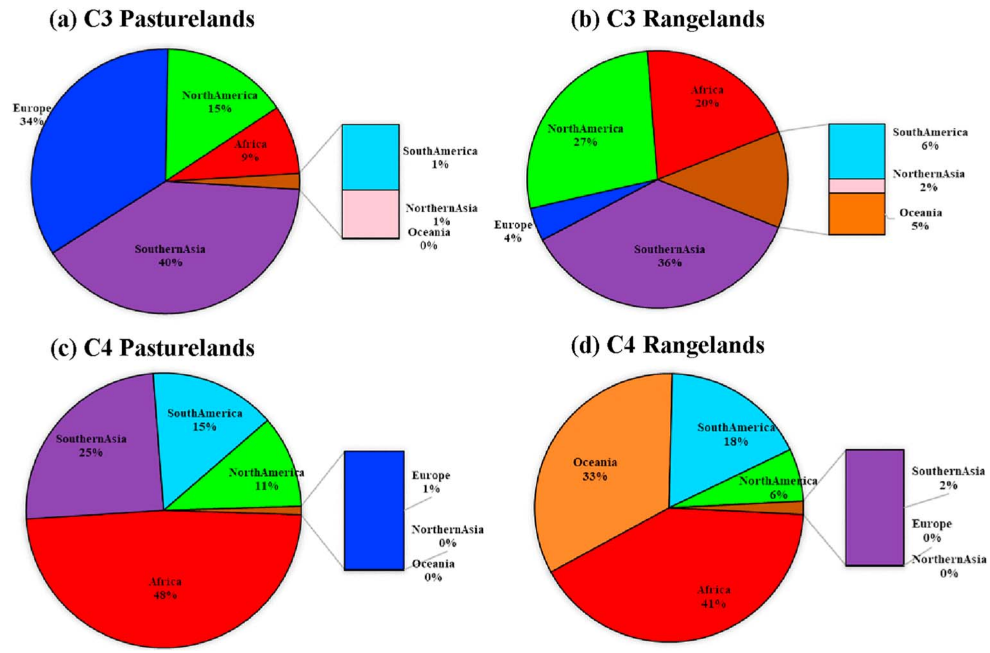

Figure 6. Regional changes in estimated N_2O emissions by different types of pasturelands and rangelands: (a) C3 pasturelands, (b) C3 rangelands, (c) C4 pasturelands, and (d) C4 rangelands.

3.4. Contemporary Changes in N_2O Emissions

During the current period (2001–2014), pasturelands and rangelands were a source of 2.9 $\pm$ 0.12 (mean $\pm$ sd) Tg N_2O-N/year, with a significant increasing trend of 0.027 Tg N_2O-N/year^2 ($p < 0.05$). In particular, the trend of pastureland emissions of 0.026 Tg N_2O-N/year^2 ($p < 0.05$) accounts for nearly all of the global trend. Rangelands only have a very small N_2O emissions trend of 0.001 Tg N_2O-N/year^2 (0.36 Tg N_2O-N/year), which was not statistically significant ($p = 0.1$). During 2001–2014, Southern Asia and Europe dominated N_2O emissions contributing to 42% and 25% of the total emissions during the contemporary period. Southern Asia experienced a significant increase in N_2O emissions at the rate of 0.015 Tg N_2O-N/year^2 ($p < 0.05$), while Europe experienced a significant decline in N_2O emissions at the rate of 0.005 Tg N_2O-N/year^2 ($p < 0.05$, Figure S7). Attribution of the mean N_2O fluxes to different sources indicated that livestock excreta deposition contributed to 51% of the total N_2O emissions, followed by manure N application (13%) and fertilizer N application (11%). Pasturelands were the dominant source contributing to 88% (2.57 Tg N_2O-N/year) of the total N_2O emissions from grassland ecosystems during 2001–2014.

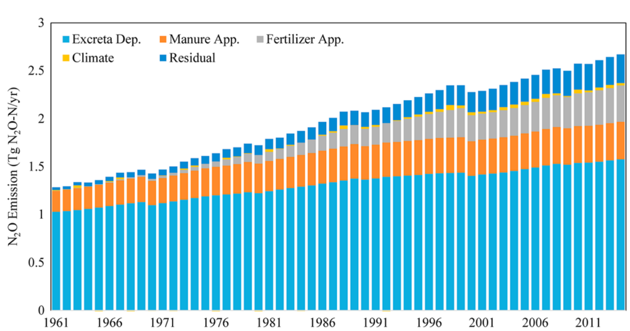

Figure 7. Attribution of N_2O emissions to excreta deposition, manure N application, N fertilizer application, and climate during 1961–2014. Preindustrial N_2O emissions based on S0 simulation were subtracted from the multifactor simulation (S1) to attribute changes in N_2O emissions to management intensity and climate.

4. Discussion

4.1. Global Nitrous Oxide Emissions and Its Comparison With Previous Studies

Over the study period (1961–2014), model simulations show that pasturelands and rangelands are responsible for a net N_2O flux of 2.4 $\pm$ 0.4 (mean $\pm$ 1 sd) Tg N_2O-N/year. Of the total N_2O fluxes, natural and anthropogenic sources accounted for 0.64 (26%) and 1.78 Tg N_2O-N/year (74%), respectively. Our study further indicates that global pasturelands and rangelands accounted for 32% (2.2 Tg N_2O-N arising from manure N, fertilizer N, and excreta N deposition for year 2006) of the total anthropogenic sources (6.9 Tg N_2O-N) based on the IPCC AR5 estimates for year 2006 (Ciais et al., 2014). When compared to global agricultural emissions (direct soil emission and emission from animal production) of about 4.1 Tg N_2O-N (Ciais et al., 2014, Table 6.9) in 2006, we found that pasturelands and rangelands are responsible for more than half (54%) of total

Table 4
Comparison of DLEM-Simulated Anthropogenic N_2O Emissions From Pasturelands and Rangelands Against Other Studies at the Global Level

SN	Methods	Year	Estimated (Tg N_2O-N)	DLEM (Tg N_2O-N)	References
1	Emission factor (animal production)	2006	2.3	1.87	Syakila and Kroeze (2011)
2	Top-down, bottom-up, linear regression	2000	2.8 (2.2–3.3)	2.0	Davidson (2009)
3	Emission factor (manure management and manure in pasture/range)	1970–2012	1.24	1.86	EDGAR (2018)
4	Process based model (ORCHIDEE)	1961–2012	1.24	1.76	Chang et al. (2016)

Note. Syakila and Kroeze (2011, (SN. 1) estimated N_2O emissions associated with animal waste management systems (manure and excreta N) using updated livestock number from FAOSTAT, which was compared with DLEM-simulated N_2O fluxes from manure N application and excreta N deposition. Davidson (2009, SN. 2) estimates N_2O emissions by fitting a multiple linear regression as a function of N input, which includes manure application to soils and livestock production. But the DLEM does not account for manure applied to agricultural crops and only considers manure applied to pasturelands. EDGAR (2018, SN. 3) was obtained by aggregating country level N_2O emissions from manure management and manure on pasture/range/paddock. Chang et al. (2016, SN. 4) provide estimates of human-induced N_2O emissions by including livestock excreta deposition (simulated internally) and manure/fertilizer application in ORCHIDEE model. DLEM = Dynamic Land Ecosystem Model; EDGAR = Emission Database for Global Atmospheric Research; FAOSTAT = Food and Agriculture Organization Corporate Statistical Database.

agricultural N_2O emissions. C3 pasturelands, in particular, were the single largest contributor to N_2O fluxes, accounting for 82% of the total N_2O emissions. We further compared the results of this study with Syakila and Kroeze (2011), who attributed total N_2O emission to direct soil emissions (1.8 Tg N_2O-N) and emission from animal production (2.3 Tg N_2O-N) in 2006. Syakila and Kroeze (2011) estimated N_2O fluxes from animal waste management systems, which consider excreta and manure N but exclude fertilizer N application. Comparison of DLEM-simulated N_2O emissions from excreta N deposition and N manure application against Syakila and Kroeze (2011) showed that DLEM underestimated N_2O emissions by 19% (0.43 Tg N_2O-N) (Table 4). Likewise, Emission Database for Global Atmospheric Research (EDGAR v4.3.2; http://edgar.jrc.ec.europa.eu/overview.php?v=42FT2010; accessed 1 May 2018) reported that manure management and manure in pasture/range/paddocks resulted in a net N_2O emissions of 1.24 Tg N_2O-N/year compared to our estimate of 1.86 Tg N_2O-N/year during 1970–2012. Our estimates of N_2O emissions was higher than EDGAR by 50%, resulting in the largest discrepancies in emissions from manure management and manure in pasture/range/paddocks. Lower estimates of N_2O emissions based on EDGAR is not surprising because EDGAR show low net N_2O emissions compared to other studies, particularly from manure management (see Table 1; Davidson & Kanter, 2014). For example, manure management related-N_2O emissions according to EDGAR was lower than FAOSTAT and Environmental Protection Agency (EPA) estimates by 33% and 50%, respectively. Although FAOSTAT, EPA, and EDGAR use IPCC tier I approach to provide estimates of N_2O emissions from manure management, the large discrepancy is because EDGAR database uses a combination of private and public data to provide estimates of N_2O emissions from different sources.

We also compared our results with two other sources that rely on statistical and process-based models. By combining top-down analysis of atmospheric N_2O accumulation constrained by bottom-up approaches and fitted parameters based on multiple linear regressions, Davidson (2009) estimated a net N_2O flux of 2.8 (2.2–3.3) Tg N_2O-N associated with manure application in soils and livestock production in 2000. DLEM simulations, however, show that N inputs in the form of manure application, fertilizer application, and excreta deposition (estimated as a difference between simulation S2 and S3) resulted in a net N_2O emissions of 2 Tg N_2O-N in 2000 (Table 4). The difference in N_2O flux of 0.8 Tg N_2O-N/year is because Davidson (2009) included emissions from manure applied to soils, which consists of both pastures and croplands; however, we only considered manure N application in pasturelands. Assuming an emission factor of 2% from manure applied to soils based on Davidson (2009) and total manure applied to croplands of about 24.5 Tg N (Zhang, Tian, et al., 2017), N_2O emissions from manure applied to croplands roughly corresponds to 0.5 Tg N_2O-N. This would result in a net N_2O flux due to manure application in soils and animal production of 2.5 Tg N_2O-N, which is 10% lower than that published by Davidson (2009), but is within the uncertainty range of 2.2–3.3 Tg N_2O-N. Using a statistical model that links N_2O emissions to soils, climate, and land use change in grassland ecosystems that receive only manure/fertilizer N, Stehfest and Bouwman (2006) estimated total (natural and human induced) N_2O emissions of about 0.81 Tg N_2O-N, which is comparable to the DLEM-estimated N_2O emissions from manure/fertilizer N application of about 0.89 Tg N_2O-N from pasturelands and rangelands. The DLEM simulations tend to overestimate N_2O emissions by 10% compared to Stehfest and Bouwman (2006), largely because of slightly higher N input in the form of manure/fertilizer N in this study. For example, total N input in the form of fertilizer/manure N in the DLEM is higher than Stehfest and Bouwman (2006) by 3.4 Tg N (see Table S2). Similarly, using a combination of process-based model (ORCHIDEE) and an emission factor, Chang et al. (2016) estimated human-induced N_2O emissions of 1.24 Tg N_2O-N/year during 1961–2012, which is lower than DLEM-simulated anthropogenic N_2O emissions (1.76 Tg N_2O-N/year) by 30% (Figure S8). The overestimation compared to Chang et al. (2016) is likely due to uncertainty associated with how livestock excreta N deposition and manure N application are handled within the model. For instance, ORCHIDEE simulates livestock excreta deposition internally in the model as a function of livestock density and available grassland forage but then uses manure application in grasslands as input into the model. The differences in the way manure deposition and application are factored into the model likely explains the slight overestimation compared to ORCHIDEE. Likewise, in some grids, the grassland productivity from ORCHIDEE for domestic animals is not able to fulfill the grass biomass use by livestock. The grassland biomass deficit led to consumption of lower grass biomass by livestock, resulting in lower excretion in the form of manure and urine from domestic animals leading to lower N_2O emissions (Chang et al., 2015, 2016). In addition, unrealistic fraction of grass and other feedstuff associated with different land cover maps for some regions may likely explain the differences in N_2O emissions.

In particular, our modeling study indicates that there are differences in the net N_2O flux from pasturelands and rangelands when compared to estimates based on emission factors. For example, studies that use default emission factor assume a linear increase in N_2O emissions as a function of fertilizer/manure N application but do not account for the effect of soil conditions, climate, and vegetation type, which varies considerably across different regions (Philibert et al., 2012; Stehfest & Bouwman, 2006). Recent studies have contended that fertilizer/manure inputs lead to linear, exponential, or hyperbolic response of soil N_2O emissions to N applications (Bouwman et al., 2002; Hoben et al., 2011; Shcherbak et al., 2014; Zebarth et al., 2008; Zhou et al., 2017), depending on whether the soil substrate is N limited or carbon limited (Kim et al., 2013). For example, in a recent meta-analysis study, Shcherbak et al. (2014) found that N_2O fluxes increased significantly faster compared to linear responses following mineral N application for most crop types. This implies that using a default emission factor for global extrapolations of N_2O emissions as a function of N inputs in the form of manure/fertilizer N might not accurately capture the biological thresholds that occur when N inputs exceed plant N demand. In the DLEM, N_2O fluxes can be lower when plant N demand exceed N inputs, particularly in systems that are N limited, provided that other factors are not limiting (Lu et al., 2012). But as N inputs exceed N demand, excess N could ultimately lead to N saturation (Fisher et al., 2007) intensifying nitrification and denitrification rates resulting in an increase in N_2O fluxes (Zhou et al., 2017). The thresholds at which these transitions occur in the DLEM largely depend on the prevalent PFTs, local climatic conditions, soil properties, land use history, and the N limitation at the site, which regulate soil microbial activities necessary for N cycling and transformation within the soil. Also, the emission factor (0.02 Tg N_2O-N/Tg N) estimated by the DLEM is higher than other emission factor-based approach, which can be explained by two possible reasons: (1) emission factors are estimated based on local studies, which are then upscaled using local emission factor and national or global N input data sets; however, such extrapolation does not account for the impact of changes in climate and soil conditions that affect N_2O fluxes at subdaily to daily timescale; and (2) the emission factor based on the DLEM was estimated by aggregating for all forms of N inputs as well as N legacy effect.

4.2. Variations in N_2O Emissions From Pasturelands and Rangelands During 1961–2014

Our results indicate that pasturelands and rangelands are an important source of N_2O with estimated global N_2O emissions of 3.11 Tg N_2O-N in 2014, and large spatial patterns and biome-specific variations. Pasturelands and rangelands are considered an important source of N_2O (Dobbie & Smith, 2003; Mosier et al., 1991), but the spatial pattern and the magnitude and sources of N_2O emissions are still debated (Davidson, 2009; Reay et al., 2012). Our results indicate that pasturelands alone contributed to 86% of the global total emissions. It is important to recognize that pasturelands only cover 24% of the total area (Klein Goldewijk et al., 2017) but contribute to 86% of global N_2O emissions from pasturelands and

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Table 5
Regional Changes in N Input and Net N_2O Emissions in Pasturelands and Rangelands During 1961–2014

Regions	Excreta N (Tg N/year)	Manure N (Tg N/year)	Fertilizer N (Tg N/year)	N_2O emissions (Tg N_2O-N/year)
Africa	15.12	0.33	0.02	0.29
Europe	5.59	2.85	1.72	0.70
North America	6.54	0.76	0.80	0.39
Northern Asia	0.26	0.12	0.01	0.02
Oceania	5.57	0.04	0.04	0.03
South America	12.50	1.07	0.09	0.06
Southern Asia	21.25	2.02	1.46	0.91
Global	2.4 Tg N_2O-N/year			
Global (anthropogenic)	1.78 Tg N_2O-N/year			
Global emission factor[a]	0.02 Tg N_2O-N/Tg N			

[a]The net N_2O emissions in fifth column is based on multifactor (S1) simulation. Emission factor was estimated as the ratio of anthropogenic N_2O emissions (difference between multifactor simulation [S1] and the simulation in the absence of external N input [S2]) and total N input in the form of excreta deposition, manure N application, and fertilizer N application.

rangelands. Higher N_2O emissions per unit of land area in pasturelands are likely due to higher N input to soils in the form of livestock excreta deposition and manure/fertilizer N application compared to rangelands (Table S3). In the DLEM, N input in the form of livestock excreta deposition, fertilizer, and manure N in pasturelands increased substrate availability (NH_4^+ and NO_3^-) due to an increase in nitrification and denitrification rates, which stimulated N_2O emissions. In particular, pasturelands are often less water limited that together with warmer climatic conditions, resulted in an overall increase in N_2O emissions.

Regionally, our results indicate that Southern Asia, Europe, and North America are the largest source of N_2O emissions during 1961–2014 (Figure 5). Higher N_2O emissions in Southern Asia, Europe, and North America are largely due to higher N input in the form of livestock excreta deposition, manure, and fertilizer N application on (Table 5). In Europe, pasturelands are commonly used as fodder or hay/silage for ruminants during the growing and nongrowing seasons (Flechard et al., 2007). As N is often limited in grassland ecosystems (LeBauer & Treseder, 2008), fertilizer N and manure N are applied to increase grassland productivity. However, application of manure/fertilizer often exceeds plant N demand, which in turn increases leaching of nitrates and gaseous emissions in the form of ammonia (NH_3), nitric oxide (NO), and N_2O (Soussana et al., 2007). In European grasslands, our results indicate that there was a significant increase in N_2O emissions until 1990, but a significant decline after 1990. The increase in N_2O emissions was due to higher N input in the form of excreta from livestock until 1990. After 1990, declining livestock numbers in Europe reduced excreta N production (Dangal, Tian, Zhang, et al., 2017), which ultimately led to low livestock excreta deposition resulting in an overall reduction in N_2O emissions. Livestock excreta deposition in Europe increased at a rate of 0.02 Tg N/year^2 during 1961–1990 but then declined at a rate of 0.11 Tg N/year^2 during 1991–2014 (FAOSTAT, 2018). The livestock excreta deposition trend in Europe based on FAOSTAT (2018) is similar to the DLEM estimated excreta deposition rate of +0.02 Tg N/year^2 until 1990 and −0.11 Tg N/year^2 after 1990.

Unlike Europe, N_2O emissions in Southern Asia increased significantly at the rate of 0.015 Tg N_2O-N/year^2 during 1961–2014. In Asia, per capita meat consumption has nearly tripled from 11 to 32 kg/year between 1961 and 2015 (FAOSTAT, 2018). In particular, China has shown dramatic increases in per capita meat consumption including pork, beef, mutton, and poultry from 12 in 1985 to 34.7 kg in 2009 (Zhou et al., 2012). Traditionally, Asia and other developing regions were dominated by extensive production units, which largely relied on local fodder, crop residues, unconsumed human food, and natural pastures to meet the demand of milk and meat productions (Steinfeld, Wassenaar, & Jutzi, 2006). However, there has been a rapid increase in animal numbers, their carcass weight (weight of an animal after being partially butchered), and per capita meat production in Asia, particularly China over the last few decades largely due to rapid population growth, urbanization, rising incomes, and dietary changes (Tilman et al., 2002). For example, cattle and buffalo populations have increased by 1.5-fold from 200 million in 1961 to 428 million in 2014 in Southern Asia, while carcass weight of cattle has increased by 1.67 times since 1961 (FAOSTAT, 2018).

Similarly, the pig population and carcass weight increased 5.3- and 1.77-fold, respectively, in China, which covers the largest part of South Asia's area (FAOSTAT, 2018). The increase in both population of livestock and their carcass weight is due to the emergence of large intensive livestock production system units in Asia, which led to an increase in N_2O emissions associated with an increase in total excretal N production (Figure 5).

Our results also suggest that pasturelands and rangelands of Southern Asia are the dominant source of N_2O emissions, contributing to 38% of the global total. The increasing contribution to N_2O emissions from Southern Asia is because of a significant increase in livestock excreta deposition, manure N, and fertilizer N application in pasturelands and rangelands ($p < 0.05$). In contrast, livestock excreta deposition and manure application have declined significantly in Europe ($p < 0.05$), while fertilizer N has increased significantly at the rate of 0.04 Tg N/year^2 ($p < 0.05$; $R^2 = 0.56$) during 1961–2014. The increase in fertilizer N and a decrease in manure N and livestock excreta deposition led to an overall significant increase in N_2O emissions at the rate of 0.004 Tg N_2O-N/year^2 ($p < 0.05$; $R^2 = 0.45$). For example, although livestock excreta N deposition and manure N application declined by 2.2 (36%) and 0.66 Tg N/year (24%), fertilizer N application increased by 1.73 Tg N/year between the 1960s and the 2010s in Europe. Likewise, our results indicate that excreta N deposition remained stable between 1961–1975 but declined significantly after 1975 ($p < 0.05$) in North America. However, manure and fertilizer N application have increased significantly ($p < 0.05$), which resulted in an increase in N_2O emissions at the rate of 0.006 and 0.036 Tg N_2O-N/year^2, respectively, since 1961 in North America.

Across different biome types, model simulations indicate that C3 pasturelands accounted for 82% of the net N_2O emissions, followed by C3 rangelands (11%), C4 pasturelands (4%), and C4 rangelands (3%, Figure 6 and Table S4). When normalized by area, pasturelands and rangelands resulted in a mean emission of 2.8 and 0.13 Kg N_2O-N/ha/year, respectively, indicating that both total emission and emission per unit area are higher in pasturelands. The large differences in emissions across four biome types can be explained by two possible reasons: (1) higher management intensity in C3 pasturelands due to additional N inputs in the form of manure/fertilizer N (Table S3) and (2) warmer and wetter climatic conditions in pasturelands indirectly modify soil temperature and soil water availability leading to higher N_2O fluxes in C3 pasturelands compared to C3 rangelands. Hartman and Niklaus (2012) found that annual N_2O emissions were higher by several orders of magnitude in intensively managed compared to extensively managed grasslands, although growing season N_2O emissions were quite similar at both sites. The differences were largely attributed to high management intensity, low N limitation, and limited plant growth responses, likely resulting in larger proportion of applied N in the soil and further intensifying nitrification and denitrification rates. In a similar study with no N limitation, N_2O fluxes showed an exponential relationship with WFPS and temperature in grassland ecosystems (Flechard et al., 2007; Smith et al., 1998). It is likely that higher management intensity and differences in prevalent climatic conditions led to higher N_2O emissions from pasturelands compared to rangelands. Comparison of air temperature and precipitation changes with total N_2O emissions estimated as a difference between S1 and S2 scenarios (see Table 1) show that the interaction of climate with external N input led to a large increase in N_2O emissions. Our analyses indicate that pasturelands have higher mean annual precipitation (1043 mm/year) compared to rangelands (490 mm/year), but the long-term trend of annual precipitation totals show no substantial change for both pasturelands and rangelands (Figure S9). Interestingly, our results indicate that pasturelands experienced a significant increase in air temperature at the rate of 0.04°C/year ($p < 0.05$; $R^2 = 0.90$). In contrast, air temperature in rangelands increased marginally at the rate of 0.007°C/year ($p < 0.05$; $R^2 = 0.19$, Figure S9). Given that precipitation did not change substantially in both pasturelands and rangelands, higher N_2O emissions from pasturelands were driven by a significant increase in air temperature (Figure S10). As pasturelands soils are less water limited due to higher precipitation compared to rangelands, an increase in temperature enhances microbial activity (Scanlon & Kiley, 2003) and possibly intensifies denitrification rates by favoring anaerobic condition, resulting in higher N_2O emissions. For example, in a grazed grasslands under different management systems, Rafique et al. (2011) found that N_2O emissions were 5 times greater at air temperature of 17°C compared to 5°C. Other studies have also reported a profound influence of temperature on N_2O fluxes between 5 and 18°C (Flechard et al., 2007; Saggar et al., 2004). Likewise, precipitation can influence N_2O emissions in two possible ways: (1) high rainfall can increase WFPS resulting in higher N_2O emissions, driven by increasing contribution from denitrification (Bateman & Baggs, 2005; Dobbie & Smith, 2001); (2) high rainfall also increases nitrate leaching, which ultimately lead to a reduction in the concentration of nitrates necessary for denitrification (Saggar et al., 2007). Davidson (1993) indicates that nitrification was the dominant source of both N_2O and NO emissions when WFPS was less than 30–60%, while denitrification was the major source of N_2O emissions when WFPS was greater than 60–80%. Higher soil moisture (above or below field capacity) exert an important control on N_2O and NO emissions, with higher NO production below field capacity and higher N_2O production above field capacity (Davidson, 1991). For example, regardless of the ecosystem types, Luo et al. (2013) found that moister and warmer soil conditions led to large N_2O fluxes, explaining 50% of the temporal variations. Unlike high N_2O fluxes from moister and warmer soils, rangelands experience frequent moisture stress with large impacts on soil microbial activity (Breman & de Wit, 1983; Dijkstra et al., 2012), where water potential of about −14 MPa in mineral soils and −36 MPa in surface litter ceases biological activity (Manzoni et al., 2012). Under simulated drought conditions in two grassland sites, Hartman and Niklaus (2012) found that drought may result in larger and sustained reduction in N_2O emissions, which are estimated to be 1 to 2 orders of magnitude lower than nondrought conditions during the growing season. Interestingly, Hartman and Niklaus (2012) indicated that fertilizer addition during drought led to even a larger decrease in N_2O emissions, but such effect was counterbalanced by high N_2O emissions at moderate to high soil moisture. Our simulation results suggest that N_2O emissions in pasturelands were driven by the activity potential or the size of the microbial pool, with large N_2O fluxes when WFPS exceeds 60%, while that in rangelands was regulated by environmental limitations, with low N_2O fluxes under conditions of moisture stress regardless of the amount of excreta N deposition.

4.3. Sources of N_2O Emissions

Our results indicate that livestock excreta N deposition was the largest source of N_2O emissions, contributing to 54% of the mean N_2O fluxes during 1961–2014. This is primarily because N input in the form of excreta N deposition has increased at a rate of 0.66 Tg N/year ($p < 0.05$; $R^2 = 0.98$), while manure and fertilizer N applications have increased at the rate of 0.06 ($p < 0.05$; $R^2 = 0.90$) and 0.18 Tg N/year ($p < 0.05$; $R^2 = 0.99$). Higher N input in the form of livestock excreta deposition increased substrate availability, which further led to an increase in the activities of microbial communities (Drenovsky et al., 2004), provided that soil water availability is not limited. The increase in activities of microbial communities affects nitrification and denitrification processes with factors such as soil water content having both synergistic and antagonistic effects depending on the status of other regulating factors such as soil aeration, N availability, and N leaching (Luo et al., 2013). Additionally, although livestock excreta N deposition was the largest source of N_2O fluxes, model results show low N_2O fluxes per unit of excreta N deposition compared to fertilizer and/or manure N application. This is because N_2O flux from rangelands is relatively low, which is largely associated with low excretal N deposition per unit area compared to pasturelands. Also, temperature and moisture trends were different in pasturelands and rangelands, with warmer and drier conditions limiting N_2O fluxes in rangelands (Flechard et al., 2007; Saggar et al., 2004). While elevated temperatures can directly stimulate nitrifiers and denitrifiers that increase N_2O fluxes, drier soil conditions associated with warmer conditions in rangelands likely reduce the activity of nitrifiers and denitrifiers (Bijoor et al., 2008).

4.4. Uncertainty Estimates

Uncertainties in the DLEM simulations come from input data, model structure, and model parameters used to simulate N_2O fluxes (Crosetto & Tarantola, 2001). Quantifying these uncertainties is complex due to the variety of N forms and microbial processes that need to be considered (Butterbach-Bahl et al., 2013; Davidson, 2009). In addition, climate, soil conditions, vegetation type, and soil management practices (fertilization and manure application) can modify N_2O fluxes due to complex interactions among different environmental factors resulting in large temporal and spatial variations (Bouwman et al., 2002; Ehrhardt et al., 2018; Tian et al., 2019). One of the largest uncertainty sources for N_2O fluxes comes from management practices that includes livestock excreta N deposition in pasturelands and rangelands and manure/fertilizer N application in pasturelands (Steinfeld, Gerber, et al., 2006; Tubiello et al., 2013). There are potentially two main causes of this uncertainty. First, manure production and their partition to croplands and pasturelands differ among studies (Bouwman et al., 2013). Second, N_2O emissions vary as a function of manure types, with cattle manure yielding higher N_2O fluxes compared to sheep manure (Velthof et al., 2003). To quantify the uncertainties in several ecosystem variables associated with uncertainties in the input data, bootstrapping, Monte Carlo, and Bayesian methods are becoming increasingly popular (Tang & Zhuang, 2009; Tian et al., 2011). In this study, we used bootstrapping techniques to assess uncertainties associated with input data (excreta N deposition, manure N application, and fertilizer N application). Figures S11–S13 show the frequency distribution and Q-Q plot of the slope (relationship between N input and N_2O fluxes) and N input in the form of excreta deposition, manure application, and fertilizer application, respectively. We used 95% confidence interval from the frequency distribution of slope and N input data to show that excreta N deposition resulted in a net N_2O emissions of 1.31 ± 0.17 Tg N_2O-N/year, while manure N application and fertilizer N application resulted in a net N_2O emissions of 0.33 ± 0.05 and 0.17 ± 0.07 Tg N_2O-N/year, respectively, during 1961–2014. The uncertainty estimates are within 13%, 15%, and 42% of the mean N_2O fluxes associated with excreta N deposition, manure N application, and fertilizer N application, respectively.

Uncertainties in N_2O fluxes also come from variation in model parameters such as maximum nitrification and denitrification rates, biological N fixation rates, and adsorption coefficient of soil ammonium (NH_4^+) and nitrate (NO_3^-). During the calibration process, we determined upper and lower limits of these parameters to derive a range of N_2O emissions in response to excreta deposition, manure application, and fertilizer application, which helped to constrain the model parameters related to N_2O emissions (Xu et al., 2017). However, to accurately quantify uncertainties related to model parameters, sensitivity analysis of the response of N_2O emissions to key model parameters is required. Sensitivity analysis of some of these parameters is possible using Monte Carlo or Bayesian techniques but requires observation data from multiple sites with different levels of excreta deposition and manure/fertilizer application (Tian et al., 2011). While data on many of the ecosystem state variables such as gross primary productivity, net primary productivity, and net ecosystem exchange are becoming available through a combination of remote sensing and eddy covariance approaches (Jung et al., 2011; Xiao et al., 2011), there are limited observations of N_2O emissions with different levels of N input, particularly in pasturelands and rangelands to carry out the sensitivity analysis of key model parameters.

Similarly, other sources of uncertainties are associated with the simplification of model processes. For example, simulation was performed at a daily time step, but Brumme et al. (1999) indicate that pulses of N_2O fluxes may vary at a subdaily scale. It is therefore important to simulate the variation in N_2O fluxes at a subdaily time step to accurately account for the net N_2O emissions from grassland ecosystems. In addition, although parameters were well calibrated based on existing field observations, second-order microbial processes (for example, models that incorporate different microbial biomass) with explicit representation of nitrifying and denitrifying bacteria have not been included in this study.

Likewise, we used spatially explicit data on livestock excreta deposition and fertilization based on Xu et al. (2019), which was developed by integrating FAOSTAT database and other data sources. We have not included the wild animal excreta deposition and only considered excreta deposition associated with domestic animals. For example, extirpation of large-bodied wild mammals has resulted in a reduction in global enteric methane emissions (Smith et al., 2016). It is possible that reduction in the population of large-bodied wild mammals can also lead to a decline in global N_2O fluxes from pasturelands and rangelands, associated with low N inputs in the form of excreta deposition.

We have also not included other management practices such as mowing/cutting frequency in pasturelands (Chang et al., 2016). While some studies show that cutting frequency can stimulate N_2O fluxes (Neftel et al., 2000; Rafique et al., 2012), others have reported reduction in N_2O fluxes after cutting events (Chen et al., 1999; Kammann et al., 1998). For example, Neftel et al. (2000) found that plants N uptake slows down following cutting events, making more inorganic N available for nitrification and denitrification, resulting in higher N_2O fluxes. Similarly, Rafique et al. (2012) suggest that N_2O fluxes were higher following cutting events, and other favorable conditions such as low/high WFPS and higher soil temperature during cutting events likely increase N_2O fluxes. In contrast, Kammann et al. (1998) indicate that cutting events likely increase competition among plants for inorganic N, thereby reducing the availability of inorganic N available for nitrification and denitrification. Likewise, Chen et al. (1999) found that lower N_2O fluxes following cutting events were largely associated with the reduction in the transport of water-dissolved N_2O through transpiration. These contradictory results indicate that further experimental evidences are required to explain the dynamics of N_2O fluxes following cutting events and to include such processes in global biogeochemical models.

5. Conclusions

In this study, we have used the DLEM to estimate the N_2O fluxes from global pasturelands and rangelands and compared these fluxes with estimates based on the IPCC AR5, EDGAR, and other global studies. Our modeling study has shown that N_2O fluxes are highly variable across time and space driven by climate, management intensity, and soil conditions. N_2O emissions generally increased with management intensity but were largely modified by soil and climatic conditions as evident from large differences in fluxes between pasturelands and rangelands. Warmer and wetter climatic conditions together with high N input in the form of excreta and manure/fertilizer application were the major driver of N_2O emissions in pasturelands. In rangelands, moisture limitation possibly resulted in low N_2O emissions despite warmer climatic conditions and high N input in the form of excreta.

Furthermore, our study has shown large global and regional variations in N_2O fluxes due to multiple environmental changes and land management practices. Regionally, our study has shown that Southern Asia experienced the largest increase in N_2O emission since the 1960s associated with changes in livestock production systems, and an increase in stocking density, per capita meat consumption, and live weight over time. In addition, our results based on single factor simulation have demonstrated that excreta deposition is the largest source of N_2O emission in pasturelands and rangelands contributing to 54% of the mean N_2O emissions, followed by manure application (13%) and fertilizer application (7%) during 1961–2014.

Despite our effort to include all major processes that influence N_2O fluxes from pasturelands and rangelands, there still remains considerable uncertainty in our understanding of processes that regulate N_2O emissions such as cutting/mowing frequency, stocking rates, and the different types of manure (Boucek, 2017). To reduce uncertainties associated with N_2O emissions from pasturelands and rangelands, we need more short- and long-term experimental studies that vary across space and with different management types (mowing/cutting, grazing, etc.) and intensities (different levels of fertilizer/manure N input). In addition, as process-based models that simulate N_2O emissions at subdaily and daily time step are becoming increasingly available (Tian et al., 2018), model intercomparison of N_2O fluxes at different experimental sites (Ehrhardt et al., 2018) and application of such models at regional and global scales (Tian et al., 2019) can help to constrain global estimates of N_2O emissions and reduce uncertainties due to model structure and their internal variability. Both experimental studies and process-based model intercomparison are required not only to constrain current estimates of N_2O fluxes but also to simulate variations in future emissions in response to different climate and land use/management scenarios.

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